Ground Water Management
Book 14 of the Series

Proceedings of the Conference entitled

Petroleum Hydrocarbons and Organic Chemicals in Ground Water: Prevention,
Detection and Restoration
Eastern Regional Ground Water Issues

November 4 - 6, 1992
The Westin Galleria
Houston, Texas

Abstract

The 1992 Petroleum Hydrocarbons Conference was comprised of 3 days of technical presentations within the following topic areas:

Search for Lost Hydrocarbons
Optimization of Groundwater Remediation Systems
Development of Remediation Levels
Free Phase Hydrocarbon Transport and Fate
Recovery of Liquid Phase Contamination
Remediation of Residual Phase Contamination: Air Sparging
Remediation of Residual Phase Contamination: Vapor Extraction
Treatment of Recovered Water
Bioremediation
Factors Affecting Remediations

In addition, more than 100 leading companies in the ground water and petroleum industries participated in the Conference Exposition in which a variety of equipment and services for preventing, detecting and remediating ground water contaminated by petroleum hydrocarbons and other organic chemicals was showcased.

The meeting was sponsored by:

The American Petroleum Institute
and
The Association of Ground Water Scientists and Engineers
(a division of
the National Ground Water Association)

This bound volume is a compilation of papers that were presented at the meeting.

Materials appearing in this publication are indexed to Ground Water On-Line, the data base of the National Ground Water Information Center at (614) 761-1711.

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* Manuscript not available at time of print.
Contact the National Ground Water Information Center at
(614) 761-1711 for information.

Trilinear Plots
A Powerful New Application for Mapping
Gasoline Contamination

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Water-quality data evaluation for gasoline-contaminated sites is often limited to a summary table of the analytical data, and possibly a graph of time plotted against either total or individual benzene, toluene, ethylbenzene and total xylenes (BTEX) compounds. In some instances ratios of benzene to total xylenes, or the presence of methyl-tertiary butyl ether, have been used to interpret the relative weathering of dissolved gasolines, or to determine the leading portion of gasoline contaminant plumes. Multivariate plots have also been used to differentiate the origins of BTEX in ground water.

A powerful new application for mapping gasoline contamination has been developed through the use of trilinear plots. Variations in the chemistry of gasoline-contaminated ground water are primarily a result of the solubility and volatility of individual compounds. The ratio of each gasoline compound with respect to another varies as a function of position within a plume, and the degree of weathering of the contaminants. Plotting the ratios of three compounds having different chemical properties graphically illustrates sources, mobility and weathering of gasoline contamination at a site.

This paper presents the results of laboratory experiments and the analyses of field data which document the effectiveness of trilinear plots. Case studies presented illustrate how trilinear plots can: 1) illustrate the zonation of a gasoline plume, and 2) identify the presence of multiple sources for contaminants.

INTRODUCTION

Millions of dollars are expended each year collecting data from sites impacted by gasoline contaminants; however, little effort is applied to interpret the resulting analytical data. Much of the analytical data gathered from gasoline-contaminated sites is simply reviewed, tabulated and presented on a graph of concentration plotted against time. A thorough

evaluation of these data can provide valuable insight into the source, nature and extent of contaminants at a site.

One investigative tool which is useful involves calculating the ratios between individual gasoline components. Ratios can be used to understand the zonation of a gasoline plume, the rate of weathering at the site, or the possibility of multiple sources. For example, plume zonation is commonly demonstrated when MTBE is the only gasoline component found in ground water downgradient of a release. At some later time, benzene may reach the same downgradient location. Calculating the MTBE-to-benzene ratio helps the investigator interpret these trends. Weathering of solutions, due to processes like volatilization and biodegradation, is often evident at gasoline-release sites, where sufficient data are collected over time. The ratio of benzene to total xylenes is higher for fresh releases than after the solution has been subjected to weathering. The same holds true for the MTBE-to-total xylenes ratios at release sites. Where multiple sources are present, chemical ratios can be used to define the extent of contaminants from each.

Reviewing ratios between several components simultaneously enhances the ability to interpret analytical data. Because the volume of data available at many sites is large, interpreting changes in ratios from tabulated data is difficult. Trilinear plots not only allow the comparison of three ratios simultaneously, but also allow many data sets to be plotted on one diagram.

PREVIOUS WORK

The technique of plotting ratios of gasoline components on trilinear diagrams to interpret the analytical data gathered at gasoline contaminated sites was first introduced by Luhrs, et al. (1992). In that study, trilinear plots were used to present the results of two laboratory experiments involving three gasolines formulated with MTBE. One experiment was conducted using water solutions saturated with gasoline; the second investigated the effects of varying concentrations.

Saturated Solutions/Weathering

Saturated solutions were prepared with three gasolines having 87, 89 and 92 octane ratings, and stored in loosely covered separatory funnels. Each solution was periodically sampled over 1 week, and analyzed using EPA Method 8240 modified to include MTBE. Table 1 summarizes the analytical results. The principal difference in the chemistry for saturated solutions using these different gasolines is the amount of MTBE. There was apparent weathering of solutions prepared with 87 and 89 octane ratings, as exhibited by decreases in MTBE and benzene concentrations over the sampling period. No similar decreases were observed for toluene, ethylbenzene or total xylene concentrations for these samples. The solution prepared with 92 octane gasoline did not exhibit similar results, although MTBE did have a minor decrease in concentration during the six days.

TABLE 1

ANALYSES OF WATER SATURATED WITH GASOLINE

Octane Rating	Sample Time in Days	Benzene	Toluene	Ethylbenzene	Total Xylenes	MTBE
87	1	32,755	46,975	3,720	16,195	12,550
	2	28,565	45,240	3,725	16,650	8,475
	6	13,115	42,210	3,385	15,245	4,725
89 (3/23/92)	1	41,270	40,775	2,645	14,570	57,195
	2	41,195	55,740	3,550	21,000	45,905
	7	9,825	39,095	3,760	20,860	15,760
89 (8/11/92)	0	51,000	72,000	5,800	27,000	80,000
	1	43,000	56,000	4,300	19,000	66,000
92	0	44,500	82,500	4,360	21,500	336,000
	1	50,500	80,900	4,210	20,300	323,000
	2	45,800	82,100	3,880	19,600	305,000
	6	44,000	81,400	4,480	19,600	287,000

All results in parts per billion

Figure 1 illustrates data from table 1 on a trilinear diagram using MTBE, benzene and total xylenes (MBX) at the apices. Weathering trends for the solutions prepared using 87 and 89 octane rated gasoline are displayed as data points progressively further away from the volatile MTBE and benzene components. The data summarized on table 1 indicates decreasing concentrations for the more volatile components. The change in chemical ratios caused by weathering of these components is only evident using the trilinear diagram. Also evident on this figure is the relative octane rating of the gasoline in solution. Because MTBE is an oxygenate used to enhance gasoline octane, the solutions prepared with the higher rated gasolines are represented closer to the MTBE apex of the diagram.

These data can also be presented on trilinear diagrams using different end members which make up the triangle. The triangular diagram included on figure 2 illustrates how the same sample data plot on a benzene, toluene, total xylene (BTX) diagram. On this figure, weathering trends for the solutions prepared with 87 and 89 octane rated gasolines show the decreasing ratio of benzene to the other two components. The spatial distribution between different octane solutions is not as evident because MTBE is not considered in the BTX plot.

Solutions of Varying Concentration

The same gasolines used in the saturation experiments were used to prepare solutions with varying concentrations. Samples were again analyzed using EPA Method 8240 modified to include MTBE. Table 2 summarizes the water-quality analyses of samples collected during this experiment. As expected, the BTEX and MTBE concentrations change according to the original water-to-gasoline ratio of the solution, and the octane rating of the gasoline used.
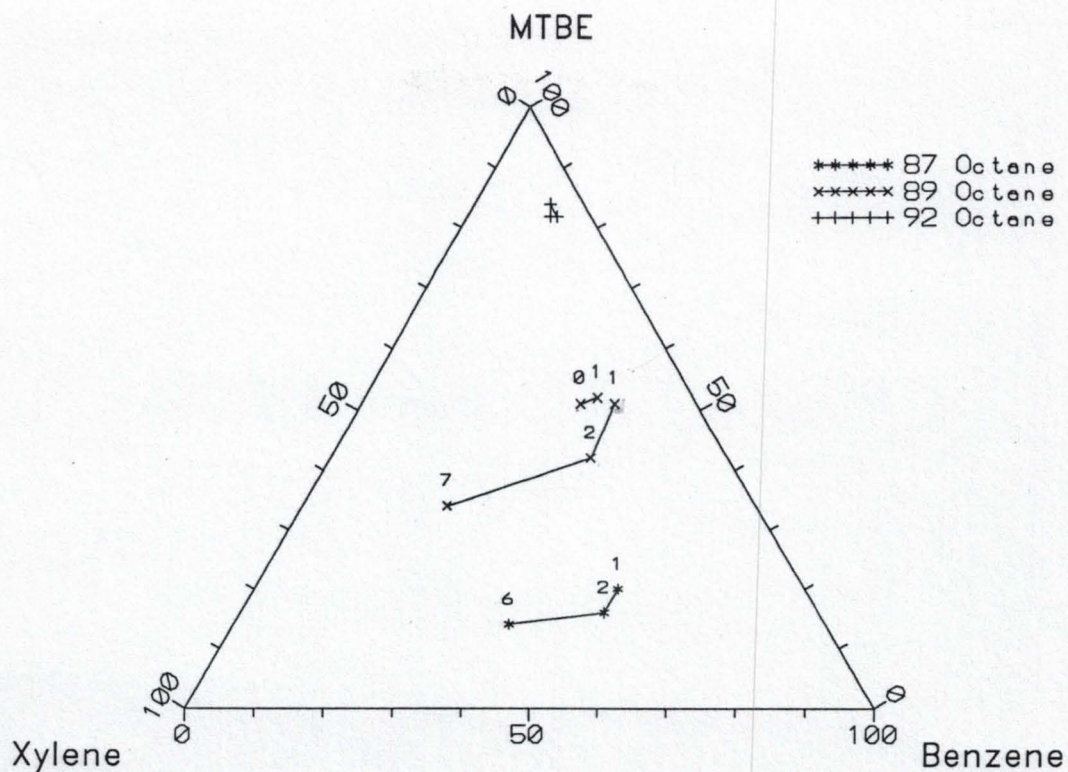
MTBE

Figure 1 - Trilinear plot of MBX ratios for data from Table 1.
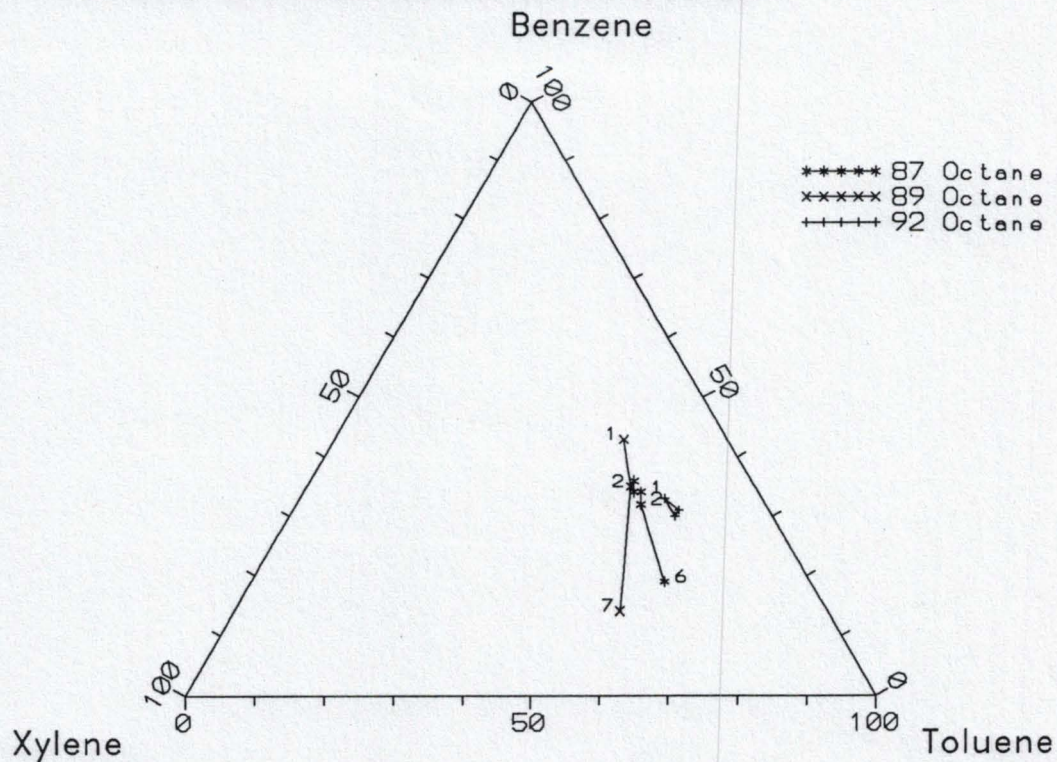
Benzene

Figure 2 - Trilinear plot of BTX ratios from Table 1.

TABLE 2

ANALYSES OF GASOLINE IN WATER SOLUTIONS

Octane Rating	ml gas per 42 ml H2O	Benzene	Toluene	Ethylbenzene	Total Xylenes	MTBE
87	1	24,000	30,000	2,400	12,000	5,300
	0.1	17,000	32,000	2,700	16,000	3,100
	0.01	1,900	7,600	1,600	7,500	85
	0.001	240	1,100	280	1,300	*9.6
89 (3/23/92)	1	36,000	35,000	2,300	13,000	20,000
	0.1	18,000	35,000	2,900	17,000	2,700
	0.01	3,100	9,200	1,700	9,000	*390
	0.001	290	1,100	260	1,500	*33
89 (8/11/92)	1	41,000	61000	4,700	22,000	38,000
	0.1	20,000	51000	4,600	22,000	4,900
	0.01	2,800	12000	2,100	9,100	500
	0.001	180	1200	360	1,600	*28
92	1	40,000	86,000	9,700	48,000	180,000
	0.1	15,000	35,000	3,700	17,000	22,000
	0.01	1,600	10,000	1,900	9,600	1,400
	0.001	88	260	120	630	99

* denotes estimated concentration below the method reporting limit
All results in parts per billion

The triangular diagram illustrated on figure 3 presents data sets from table 2 using MBX as end members. Although not readily evident on the table, figure 3 illustrates that the ratios between some components varies with the concentration of the solution prepared. Trends similar to weathering patterns are exhibited by the data. These trends are a reflection of the concentration of the solution. The proportions of individual components are in part a function of the percent of each in gasoline, its solubility in water, and partitioning. For example, total xylenes comprise nearly 10 percent of some gasolines compared to less than 2 percent for benzene (Bruce, et al. 1991). Accordingly, total xylene concentrations in solution, when no free-phase hydrocarbons are present, will be dominant until its saturation limits are reached. The spatial distribution as a function of octane is again evident because of MTBE, especially at higher concentrations.

Figure 4 presents the data from Table 2 illustrated on a BTX trilinear diagram. This plot is similar to figure 3 except for the lack of spatial distribution for solutions prepared with differing octane gasolines. The low-concentration data plot closer to the xylene corner, again because of the physical and chemical characteristics of the compound.
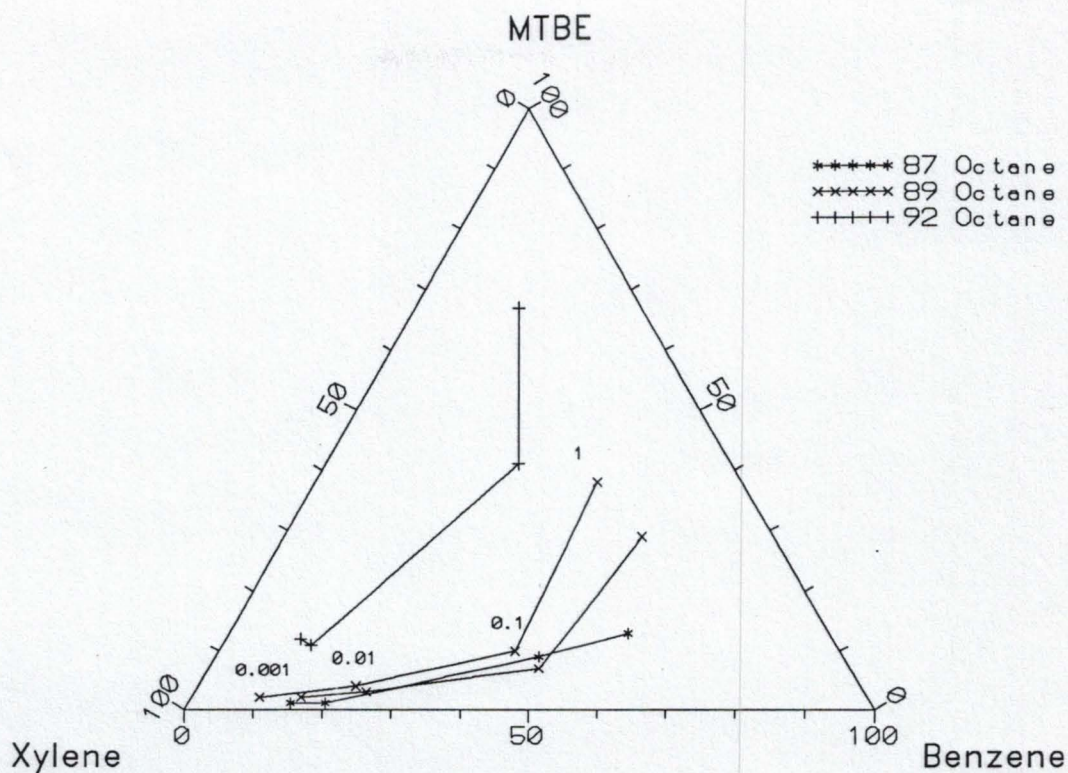
Figure 3 - Trilinear plot of MBX ratios from Table 2.
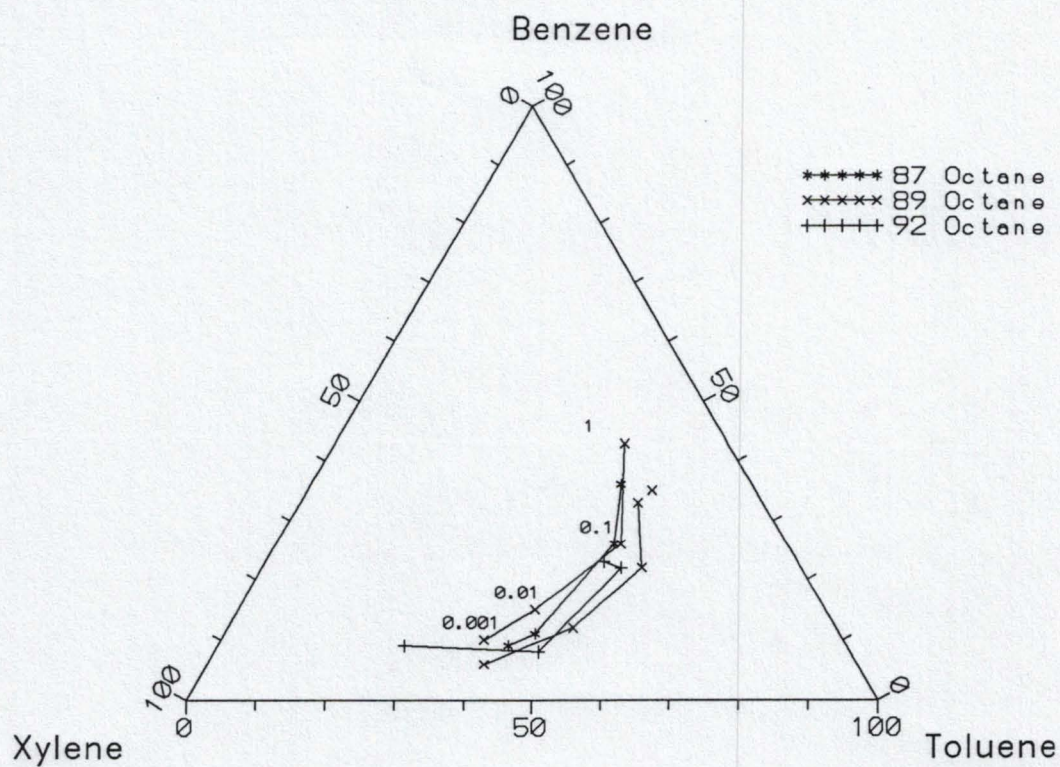
Figure 4 - Trilinear plot of BTX ratios from Table 2.

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CASE EXAMPLES

The following two case examples are presented to illustrate the benefit of using trilinear diagrams for analytical data interpretation. Each site was chosen to emphasize different information which can be gained through use of these diagrams. Both sites are in New England; however, to protect the property owners, no other geographical information is presented.

Case I

This case involves a retail gasoline site which dates back to the 1940's. The geology of the property is characterized by fill overlying a clay unit, all above a medium grade metamorphic bedrock which has been folded so the bedding planes strike in a north-south direction, and have near vertical dips. Surrounding the station are homes which, until recently, were supplied potable water by residential wells (figure 5). Currently, the underground storage tanks (UST's) are located in the southwest corner of the site. The bottom of the tank excavation is in bedrock.

The ground-water flow across the site is east-southeast, which is consistent with the regional topography. Monitor wells installed on the site and on surrounding properties represent both the overburden and bedrock ground-water systems. Results from water-quality analyses are summarized in table 3.

Figure 6 is a BTX trilinear diagram of the data contained in table 3. At first glance, the data appear to be randomly scattered across much of the trilinear plot. This scatter is not random, but a reflection of plume zonation. The data for overburden wells in the UST and dispenser island area appear in the xylenes third of the diagram. This indicates some weathering of the dissolved gasoline. Overburden wells MW-5 and MW-6 located downgradient of the tanks have similar chemistry (table 3).

Six bedrock wells located down and crossgradient from the site all have contaminants indicative of plume halo effects. Figure 6 demonstrates that all but one bedrock well (B-1) plot along the benzene-toluene axis. All these wells have similar chemistry, comprised of at least 75 percent benzene. Two bedrock wells, B-2 and B-9, represent the most cross and downgradient portion of the plume. Of the data plotted, these wells contain only benzene, and therefore plot at the top apex of the diagram. This is an example of the zonation of contaminants which form a halo around the central portion of the plume as the more mobile components preferentially migrate. Bedrock well B-1 is located near and has chemistry similar to the source area. Because ground-water flow in the area is vertically upward, gasoline contaminants would not be expected to migrate downward. The chemistry observed in this well suggests that the pumping of this well is likely the cause of contaminants entering the bedrock aquifer.

STREAM

⊕ B-6

TEST PIT

MW-12 ⊕ B-2

⊕ B-3

B-4 ⊕

B-5 ⊕

B-7 ⊕

MW-6

B-1 ⊕

MW-5

MW-10

B-8 ⊕

B-9 ⊕

MW-4 MW-3 MW-2 MW-1

GW FLOW

GW FLOW

B-10

MW-7

92

0   50   100

APPROX. SCALE (FEET)

◆⊕ N

LEGEND

– · – ·   APPROXIMATE PROPERTY LINE

[⎯⎯]   APPROXIMATE TANK FIELD LOCATION

≡≡≡   CULVERT (18" CAST METAL PIPE)

◑   OVERBURDEN WELL

◒   OVERBURDEN WELL IN TANK FIELD

⊕   BEDROCK WELL

◐   TEST PIT

CASE 1

WELL LOCATION MAP

FIGURE 5
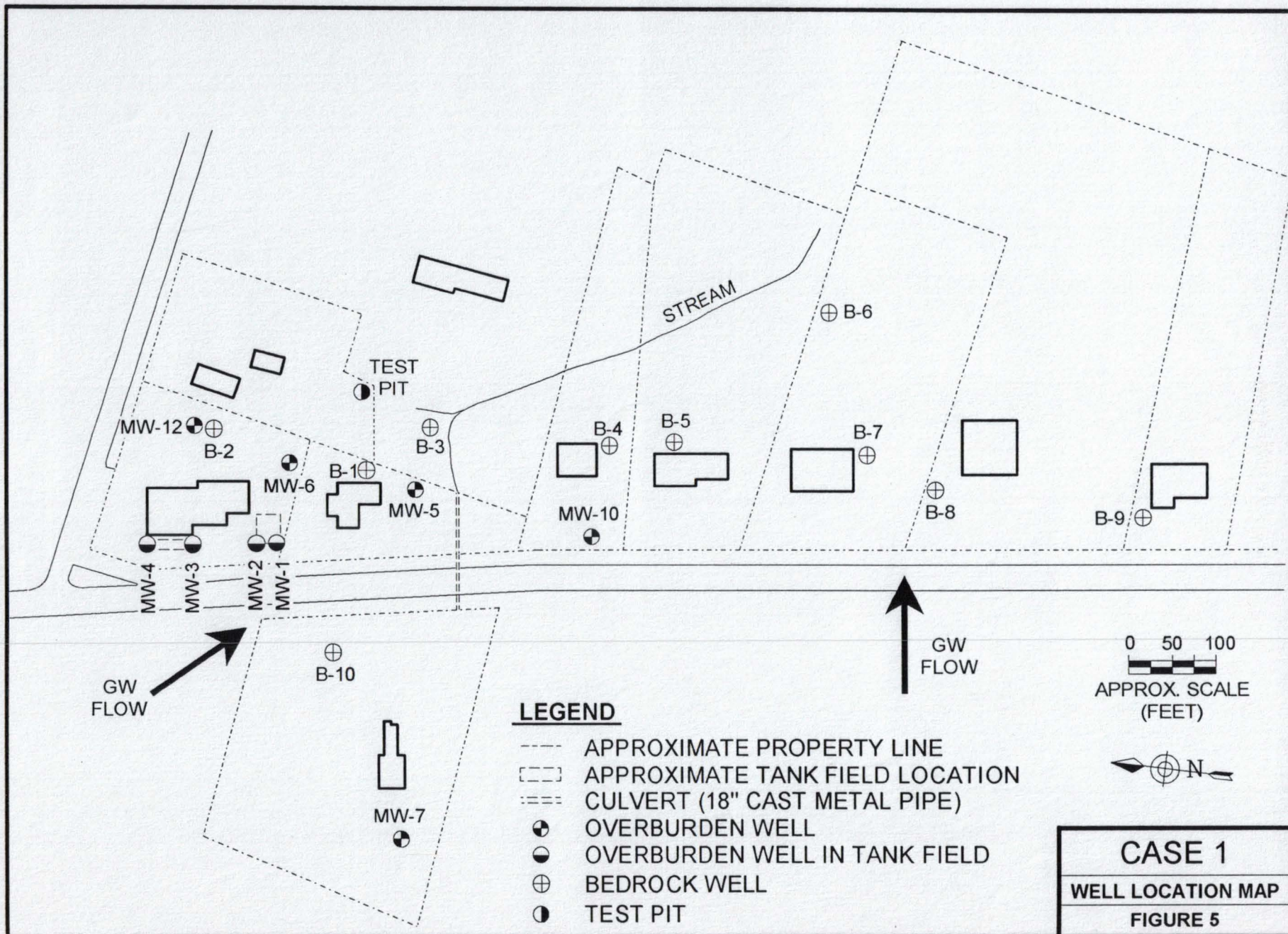
TABLE 3

WATER-QUALITY DATA - CASE I

Well ID	Benzene	Toluene	Ethylbenzene	Xylenes	MTBE
MW-1	5,700	19,000	3,300	18,000	26,000
MW-2	3,800	7,600	1,600	9,400	18,000
MW-3	33	9.3	44	90	24
MW-4	NA	NA	NA	NA	NA
MW-5	220	300	0	830	180
MW-6	12	5.9	0	8.1	31
MW-7	0	0	0	0	0
MW-10	0	0	0	0	0
MW-12	0	0	0	0	53
B-1	960	410	110	560	750
B-2	15	0	0	0	180
B-3	100	28	9.9	4.1	240
B-4	210	38	11	0	220
B-5	0	0	0	0	0
B-6	0	0	0	0	0
B-7	94	25	0	0	120
B-8	27	0	0	0	32
B-9	5.6	0	0	0	7.2
B-10	0	0	0	0	0

All results in parts per billion
NA = not analyzed
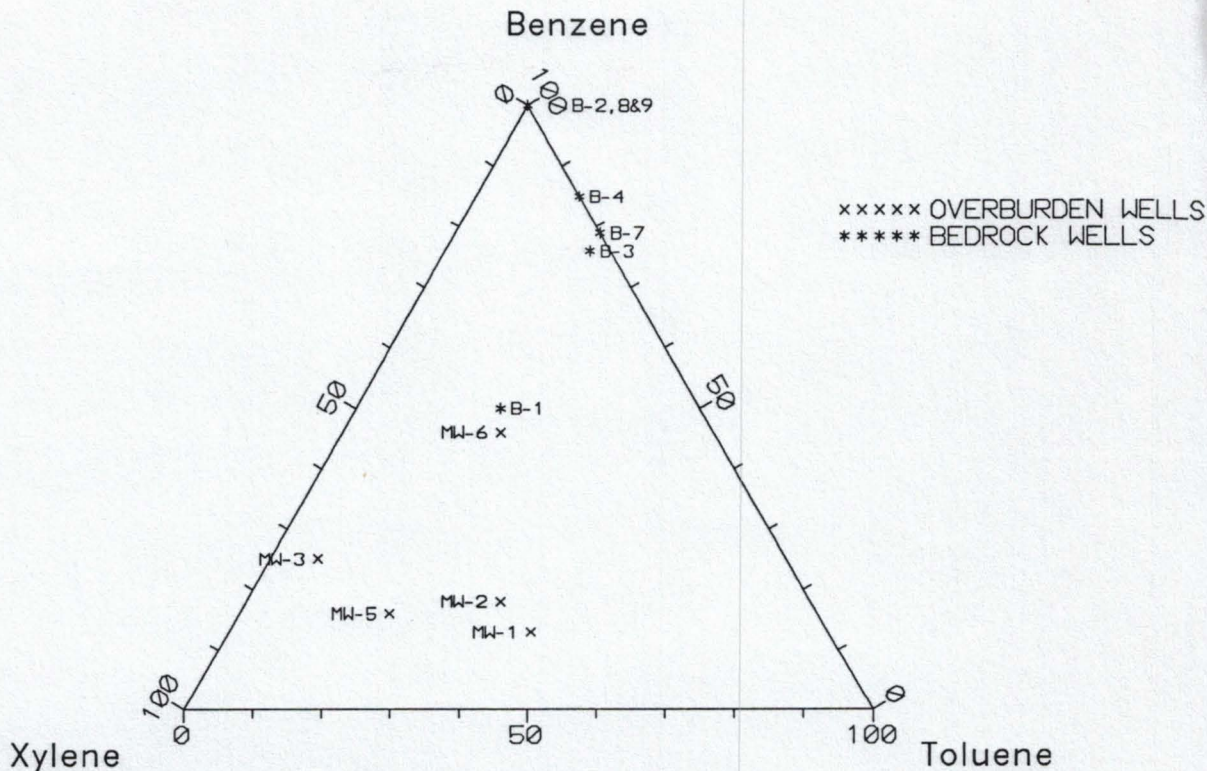
Figure 6. - BTX triangular diagram for Case I.

Case II

Case II illustrates the presence of multiple sources in one study area. Figure 7 is a site map which shows three active gasoline stations, and one former station located at the intersection of two roads. The sediments are comprised of glacial till overlying granitic bedrock at a depth of less than 20 feet. Regional ground-water flow is to the south.

Ground-water quality samples were collected from each of the wells identified on figure 7 in November, 1991. All data are summarized on table 4. Figure 8 is an MBX trilinear diagram illustrating the data on the table. Examination of this figure, in conjunction with table 4 and the site plan, indicate separate sources for the gasoline contaminants.

Two wells from Property 1 (P1-1 and P1-2) have minor concentrations of MTBE and no other dissolved hydrocarbons. Both of these wells plot at the MTBE apex of figure 8. One well located immediately downgradient on Property 2 (P2-1) also contained MTBE and no BTEX, and plots in the same location of the diagram. All other wells for both properties had non-detectable results for this sampling round.

The nature of gasoline contaminants on Property 3 is dramatically different. Figure 8 shows analytical data for wells on this property as being dominated by total xylenes. As discussed for figure 1, this is representative of an older weathered solution. Well P4-1 located

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PROPERTY 3

P3-3

PROPERTY 1

P1-3

P1-4

P3-4

P3-5 P3-1 P3-2

P1-1

P1-2

GW
FLOW

P4-1

PROPERTY 4

P2-1

P4-3

P4-2

C-1

PROPERTY 2

COMMERCIAL

P2-2

C-2

N

0   45   90

APPROX. SCALE
(FEET)

LEGEND

— · · · — APPROXIMATE
          PROPERTY LINE

DISPENSER ISLAND

MONITOR WELL

FORMER TANK LOCATION

CURRENT TANK LOCATION

CASE 2

SITE MAP

FIGURE 7

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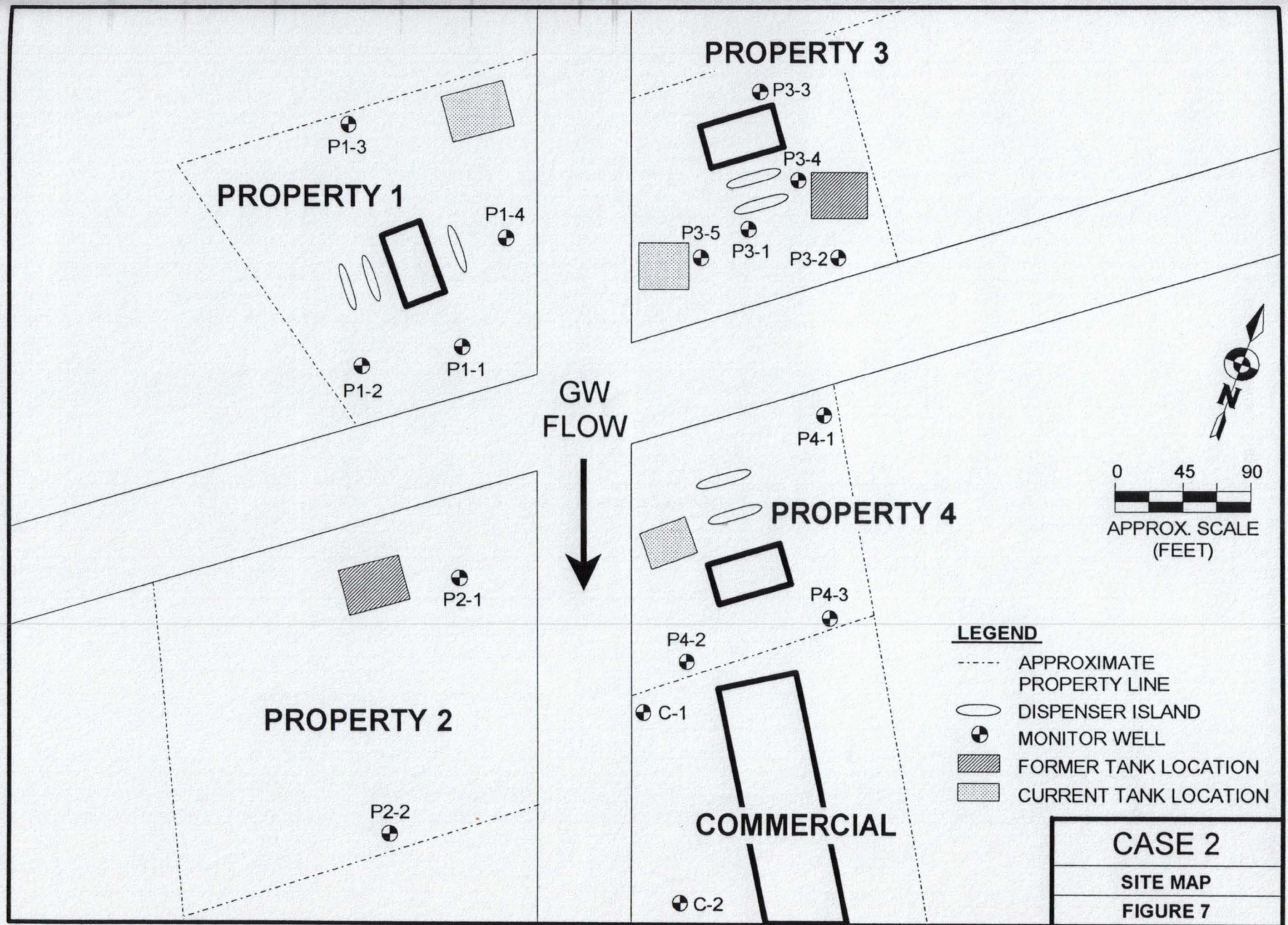
downgradient on Property 4 has similar chemistry. Positioned on the upgradient side of Property 4, this well is upgradient of any potential sources on that site (figure 7).

Dissolved hydrocarbons identified in Property 4 wells, with the exception of P4-1, indicate yet another source of gasoline contaminants. For these wells, the data plot midway along the MTBE and xylene side of the triangle. The lack of benzene in these wells is not consistent with the data presented on figure 1. Contamination of this nature is interpreted as the MTBE leading edge of a relatively fresh spill, possibly commingling with an older total xylene dominant plume.

Two wells, C-1 and C-2, exist on a commercial property downgradient of Property 4. These wells have gasoline contaminants significantly different from each other. Well C-1 plots near the xylene corner of figure 8, indicative of weathered contaminants. This well is located immediately downgradient of the Property 4 UST's. A spill from one of the UST's during 1978 is the probable source for the xylenes present in C-1. The second well (C-2) is located about 150 feet downgradient of Property 4 and plots on the MTBE corner of figure 7. This plot position is typically interpreted as the leading edge of a gasoline plume. This well data is not consistent with the data for Well C-1 which contains benzene and no MTBE, and therefore may be an indication of a local source such as a leaking vehicle.

TABLE 4

WATER-QUALITY DATA - CASE II

Well ID	Benzene	Toluene	Ethylbenzene	Total Xylenes	MTBE
Property 1					
P1-1	ND	ND	ND	ND	20
P1-2	ND	ND	ND	ND	7
P1-3	ND	ND	ND	ND	ND
P1-4	ND	ND	ND	ND	ND
Property 2					
P2-1	ND	ND	ND	ND	10
P2-2	ND	ND	ND	ND	ND
Property 3					
P3-1	ND	ND	ND	6	ND
P3-2	ND	13	ND	1500	ND
P3-3	ND	ND	ND	ND	ND
P3-4	ND	ND	ND	ND	ND
P3-5	ND	ND	ND	ND	ND
Property 4					
P4-1	ND	6	ND	1700	ND
P4-2	46	6	67	380	200
P4-3	ND	ND	ND	11	11
Commercial					
C-1	440	4,200	140	4,400	ND
C-2	ND	ND	ND	ND	180

All results in parts per billion; ND = not detected.
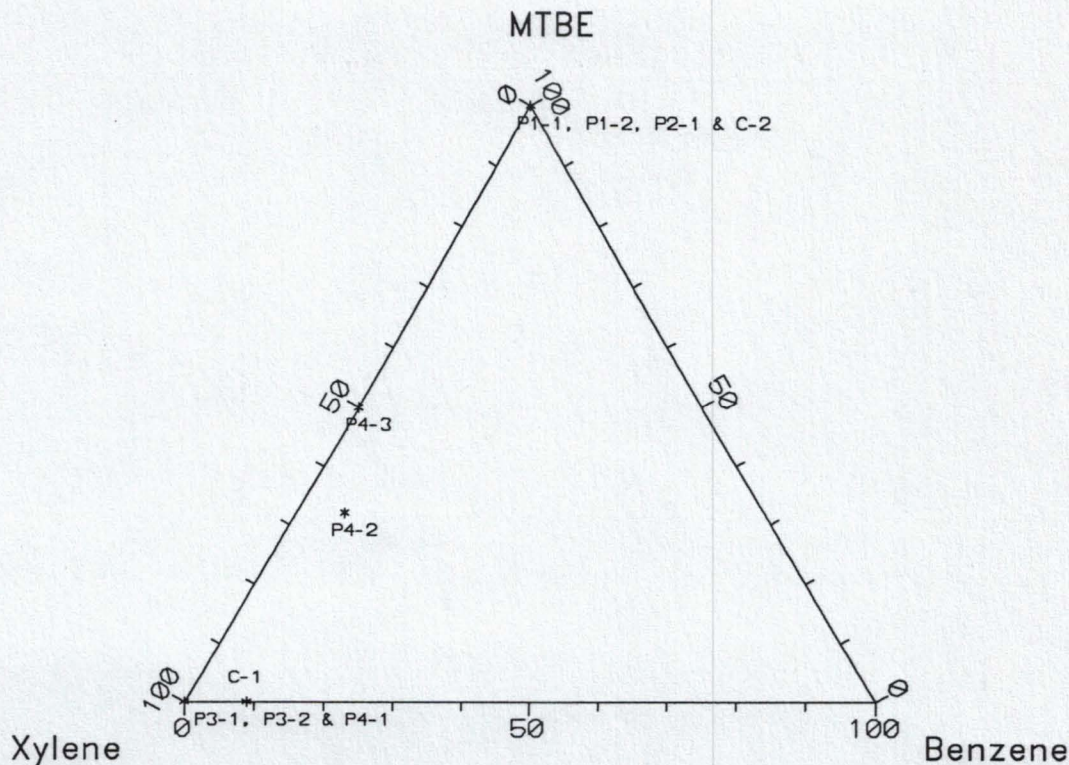
Figure 8 - MBX trilinear diagram for Case II data.

CONCLUSIONS

Trilinear diagrams prove extremely effective for graphically representing plume zonation and contaminant source identification. Without the use of these diagrams, interpretation of tabulated data is a formidable task. All trilinear diagrams should be constructed in a similar manner to facilitate interpretation. The authors have standardized the placement of the compound with the lowest boiling point at the top, and the highest at the bottom left corner of the triangle. Starting at the top of the triangle, and proceeding clockwise, the major gasoline compound order is MTBE, benzene, toluene, ethylbenzene, and total xylenes.

Case I demonstrates the zonation of a gasoline plume. Overburden wells located near the source area plot on figure 6 near the xylene corner. This is in sharp contrast to the chemistry displayed by the bedrock wells which plot close to the benzene corner of the figure. This is expected because the lighter more mobile gasoline compounds preferentially migrate away from the source area, forming halos around the central portion of the plume. Trilinear plots used to present these data clearly depict this zonation of the plume chemistry.

Case II illustrates how trilinear diagrams can be used to identify various source areas for spills. Figure 8 explicitly defines three separate source areas associated with five properties.

Data from Properties 1 and 2 plot on the MTBE corner of the diagram. Data from properties 3 and the most upgradient well of Property 4 plot on the xylene corner. The remaining Property 4 wells plot along the MTBE-xylene line. Separation of these sources is possible when the spills have relatively different ages, allowing a comparison of the current chemical composition regardless of the original gasoline fingerprint.

Conclusions which can be drawn from using trilinear diagrams include:

- **Multiple source identification**
- **The zonation of plumes**
- **Weathering of contaminants**
- **Relative octane rating of fresh spills**
- **Possible size of the release**
- **Relative age of contaminants**

Using trilinear diagrams to better understand the nature and extent of gasoline spills can be used to focus the expenditure of limited environmental budgets. Defining the presence of multiple sources is critical in evaluating responsible party liability. Where liability exists, having a clear understanding of the source, extent and zonation of contaminant plumes aids in the proper design and placement of recovery systems.

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BIOGRAPHIES

Robert C. Luhrs - Mr. Luhrs received a B.S. degree in Geology from the State University of New York at Plattsburgh in 1981. From 1981 to 1985 he worked in mineral exploration in 9 states. In 1987, Mr. Luhrs received an M.S. degree in Environmental Pollution Control at Pennsylvania State University, an interdisciplinary program including hydrogeology, ground-water geochemistry and engineering courses. Since completing graduate school, he has been employed with Leggette, Brashears & Graham, Inc. (LBG) as a hydrogeologist. His experience with LBG has been both at the company's headquarters in Wilton, Connecticut and the New England regional office in Nashua, New Hampshire, and includes management of both water-supply and ground-water contamination projects.

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ACKNOWLEDGEMENTS